Electromagnetic response of $LaO_{0.94}F_{0.06}FeAs$: AC susceptibility and microwave surface resistance

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Abstract. We discuss on the electromagnetic response of a polycrystalline sample of $LaO_{0.94}F_{0.06}FeAs$ exposed to DC magnetic fields up to 10 kOe. The low- and high-frequency responses have been investigated by measuring the AC susceptibility at 100 kHz and the microwave surface resistance at 9.6 GHz. At low as well as high DC magnetic fields, the susceptibility strongly depends on the amplitude of the AC driving field, highlighting enhanced nonlinear effects. The field dependence of the AC susceptibility exhibits a magnetic hysteresis that can be justified considering the intragrain-field-penetration effects on the intergrain critical current density. The microwave surface resistance exhibits a clockwise magnetic hysteresis, which cannot be justified in the framework of the critical-state models of the Abrikosov-fluxon lattice; it may have the same origin as that detected in the susceptibility.

1. Introduction

It is by now accepted that polycrystalline superconductors (SC) can be modeled as superconducting grains connected by weak links [1, 2, 3]. The system of intergrain contacts forms an effective pinning medium, generally indicated as intergrain region. The weak coupling of the intergrain region strongly affects the electromagnetic (em) response, especially when a DC magnetic field is superimposed to the AC driving field. Weak magnetic fields, even smaller than the lower critical field of grains, H_{c1}, easily penetrates the intergrain region through Josephson vortices. On increasing the field above H_{c1}, flux penetrates the superconducting grains and further mechanisms, of flux penetration and energy dissipation, come into play.

The granularity effects in the *in-field em* response of ceramic cuprate SC have been investigated and discussed by several authors [1, 2, 3, 4, 5, 6]. Soon after the discovery of superconductivity in iron-based pnictides, some authors have highlighted

em-granularity effects [7, 8]. In this paper, we report a study of the in-field *em* response of a polycrystalline sample of $LaO_{0.94}F_{0.06}FeAs$. We have measured the AC susceptibility, $\chi = \chi' + i\chi''$, at 100 kHz and the microwave (mw) surface resistance, R_s, at 9.6 GHz. In order to obtain information on the intergrain and/or intragrain vortex dynamics, χ and R_s has been investigated as a function of the temperature, at fixed applied fields, and as a function of the magnetic field, at fixed temperatures. At low as well as high DC magnetic fields, χ strongly depends on the amplitude of the AC field, highlighting enhanced nonlinear effects. The field dependence of χ exhibits a magnetic hysteresis that can be justified considering the intragrain-field-penetration effects on the intergrain critical current density. The field dependence of R_s exhibits a clockwise hysteresis similar to that observed in χ'; we suggest that the hysteretic behaviors of χ and R_s have the same origin and, in particular, are due to the granular nature of the sample.

2. Experimental and Sample

The *em* response has been measured in a polycrystalline sample of $LaO_{0.94}F_{0.06}FeAs$. The specimen was prepared by solid state reaction using LaAs, Fe_2O_3, Fe and LaF_3 as starting materials; details on the preparation and properties of the sample are reported in Ref. [9].

The AC susceptibility at 100 kHz has been measured by a standard two-coil susceptometer [10], coupled to an *hp*-4263 B LCR meter. The sample is located at the center of one of the secondary coils by a sapphire holder, at which a temperature sensor and a heater are fixed. The mw surface resistance has been measured by the cavity-perturbation technique [11]. A cylindrical copper cavity is tuned in the TE_{011} mode [12] resonating at 9.6 GHz; the sample is located at the center of the cavity, where the mw magnetic field is maximum.

The susceptometer (or the cavity) is placed between the poles of an electromagnet which generates DC magnetic fields up to $H_0 = 10$ kOe. Two additional coils, independently fed, allow compensating the residual field, within 0.01 Oe, and working at low magnetic fields.

3. Results and Discussion

3.1. AC Susceptibility

The real and imaginary components of the AC susceptibility at 100 kHz have been investigated as a function of the temperature (from $T = 4.2$ K up to $T \approx 30$ K), the amplitude of the AC field (up to $H_{ac} = 3$ Oe) and the DC magnetic field (at increasing and decreasing values), in the field geometry $\boldsymbol{H}_0 \parallel \boldsymbol{H}_{ac}$. Fig. 1 shows the temperature dependence of χ' and χ'', obtained in zero-field-cooled (ZFC) sample, at $H_0 = 0$ (a) and $H_0 = 5$ kOe (b) for different values of H_{ac}. The inset shows χ' and χ'' as a function of

H_{ac}, at $H_0 = 0$ and $T = 4.2$ K. All the curves are normalized to the value χ_0 obtained extrapolating the curves $|\chi'(H_{ac})|$ to $H_{ac} \to 0$, at $T = 4.2$ K and $H_0 = 0$.

From Fig. 1, whether for $H_0 = 0$ or for $H_0 = 5$ kOe, one can note a near-T_c region characterizing the linear response (χ independent of H_{ac}) and a region at lower temperatures highlighting enhanced nonlinear effects. For $H_0 = 0$, the linear response is ascribable to reversible flux penetration in the superconducting grains; the nonlinear response is due to intergrain critical state [1, 5, 14]. The peak in χ'' corresponds to the maximum energy losses, occurring when H_{ac} penetrates to the center of the sample through the intergranular region. As expected, on increasing H_{ac} the loss peak moves toward lower temperatures; however, the peak shift is much more enhanced than that observed in cuprate *SC* [5, 14].

From $\chi''(H_{ac})$ of Fig. 1 (a), the intergrain full penetration field, H_m^*, at $T = 4.2$ K results about 0.6 Oe. This value is very small, suggesting a extremely low intergrain critical current density, J_{cm}. However, we would like to remark that in this sample we have detected a strong frequency dependence of the $\chi''(T)$-peak position [15]. In particular, at 10 kHz, $H_m^*(4.2\text{ K}) \approx 0.1$ Oe. This frequency dependence indicates relaxation of the intergrain critical state with characteristic times lower than 10^{-5} s, and leads to underestimate J_{cm}.

At $H_0 > H_{c1}$, Abrikosov fluxons penetrate the grains affecting the $\chi(T)$ features. In this case, two peaks in $\chi''(T)$, and two steps in $\chi'(T)$, are expected. The low-T peak is due to full penetration of H_{ac} in the intergrain region and the high-T peak is due to full penetration of H_{ac} in the grains [1, 14]. We have performed measurements up to $H_0 = 10$ kOe, but two well distinct peaks have never been observed; this could be caused by a wide grain-size distribution, which makes the high-T peak broadened and embedded in the tail of the intergrain $\chi''(T)$ peak. Therefore, the results of Fig. 1 (b) may be justified assuming that the peak at $T \approx 9$ K is due to the intergrain full

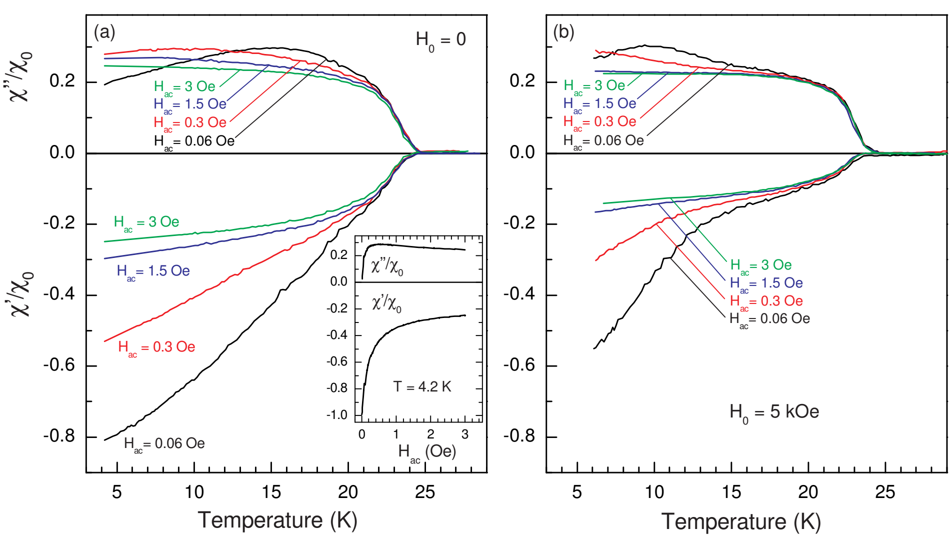

Figure 1: Real and imaginary components of the *AC* susceptibility at 100 kHz as a function of the temperature, obtained at different value of H_{ac}. (a) $H_0 = 0$; (b) $H_0 = 5$ kOe. Inset: χ' and χ'' as a function of H_{ac}, at $H_0 = 0$ and $T = 4.2$ K.

penetration of H_{ac} and the *kink* at ~ 20 K to the intragrain full penetration.

In Fig. 2, we report the DC field dependence of the AC susceptibility, obtained at $T = 4.2$ K by sweeping H_0 from 0 to H_{max} and back, for different values of H_{max}. All the curves show a reversible behavior for $H_{max} \leq 100$ Oe; when H_{max} slightly overcomes 100 Oe, a hysteresis appears. The hysteresis-loop amplitude depends on H_{ac}. As one can see, for $H_{ac} = 1.5$ Oe no hysteresis is observed at $H_0 \gtrsim 5$ kOe; on the contrary, for $H_{ac} = 0.3$ Oe the hysteresis in χ' is noticeable in the whole range of DC fields investigated. We do not report the field dependence of χ'' obtained at $H_{ac} = 0.3$ Oe because the signal is very weak and the curve is very noisy; however, we would like to remark that at low AC fields the hysteresis in χ'' reduces. It is worth noting that the increasing-field branch of the $\chi''(H_0)$ curve obtained at $H_{ac} = 1.5$ Oe exhibits a monotonic decrease, this occurs because $H_{ac} > H_m^*$. Measurements performed at $H_{ac} < H_m^*$ have shown an initial rise and a following decrease, as expected.

The hysteretic behavior of $\chi(H_0)$ can be explained by the models discussed in [2, 4, 6], in the framework of which the hysteresis in χ is related to the hysteresis of J_{cm}. The magnetic history of J_{cm} can be qualitatively described as follows. On increasing H_0 from the ZFC condition, the current through the intergrain region decreases according to the Josephson-junction behavior; a further reduction occurs when H_0 reaches the first-penetration field of grains, because Abrikosov-fluxon gradient inside the grains produces a diamagnetic moment. On decreasing H_0 from H_{max}, the moment of grains becomes paramagnetic and reduces the average effective field at the junctions; this makes $J_{cm}(H_0)$

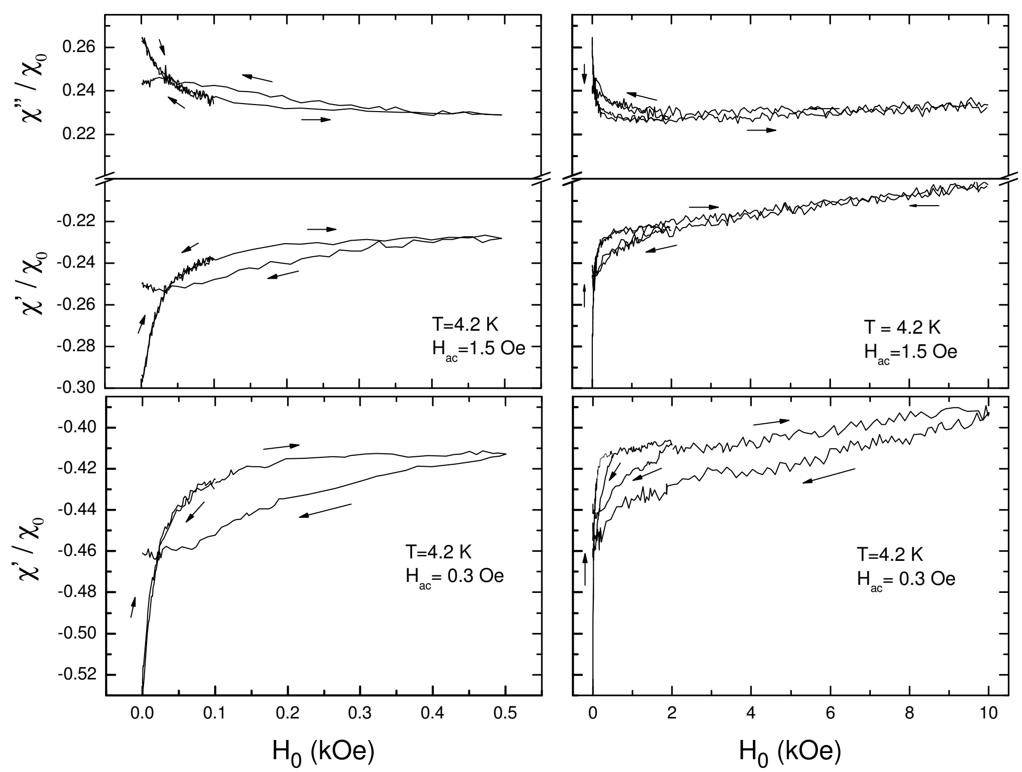

Figure 2: χ' and χ'' as a function of the DC magnetic field, obtained sweeping H_0 from 0 to H_{max} and back, for different values of H_{max}.

larger. When H_0 reaches the value of the paramagnetic contribution, the decreasing-field branch of the $J_{cm}(H_0)$ curve shows a peak, whose position depends on H_{max} and shifts toward higher fields when H_{max} increases. According to these considerations and looking at Fig. 2, we infer that the first-penetration field of grains is ≈ 100 Oe ($\chi(H_0)$ is reversible for $H_{max} \leq 100$ Oe). Moreover, the clockwise hysteresis of $\chi'(H_0)$ and the counterclockwise one of $\chi''(H_0)$, as well as their dependence on H_{ac}, can be justified in the framework of the cited models.

We have investigated the field dependence of χ at different temperatures, from 4.2 K up to T_c. On increasing the temperature, the amplitude of the hysteresis loop decreases and the range of H_0 in which it is detectable shrinks; eventually, at $T \approx 20$ K the $\chi(H_0)$ curve exhibits a reversible behavior in the whole range of fields investigated. This result is consistent with those of Fig. 1, in which a linear response is detected at $T \approx 20$ K, which is ascribable to the intragrain contribution.

3.2. Microwave Surface Resistance

The *mw* surface resistance, R_s, has been measured in the *ZFC* $LaO_{0.94}F_{0.06}FeAs$ sample as a function of the temperature, at fixed H_0 values, and as a function of H_0 at fixed temperatures, in the field geometry $\boldsymbol{H}_0 \perp \boldsymbol{H}_{ac}$. The measurements have been performed at very low input power; the estimated amplitude of the *mw* magnetic field is of the order of 1 mOe.

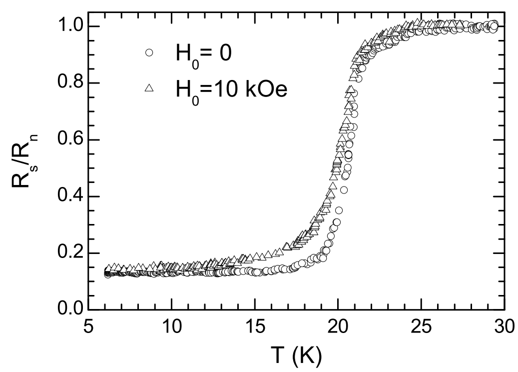

Figure 3: R_s/R_n as a function of temperature in the *ZFC* sample, at $H_0 = 0$ and $H_0 = 10$ kOe.

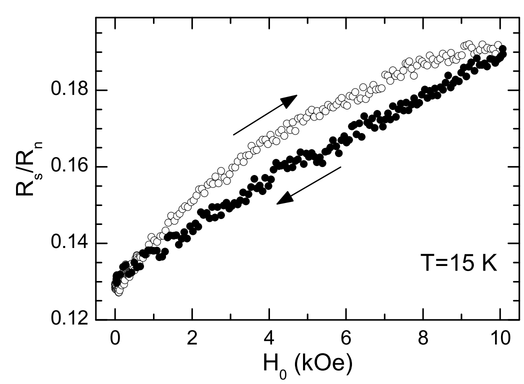

Figure 4: R_s/R_n as a function of H_0, obtained by sweeping the field up to 10 kOe and back.

Fig. 3 shows the temperature dependence of R_s/R_n obtained at $H_0 = 0$ and $H_0 = 10$ kOe; R_n is the *mw* surface resistance at $T \gtrsim T_c$. Considering the geometry factor of the sample, we estimate $R_n \approx 0.5\ \Omega$ and the residual *mw* surface resistance, at $T = 4.2$ K and $H_0 = 0$, $R_{res} \approx 65$ mΩ; the value of R_{res} is very high and confirms that a large number of weak links contribute to the *mw* energy dissipation. As one can see, the variation of R_s induced by a magnetic field of 10 kOe is very low up to temperatures $\lesssim T_c/2$ most likely because of the high upper-critical-field value of these *SC*; this is consistent with the not-visible shift of T_c with H_0.

Fig. 4 shows the normalized field-induced variations of R_s of the *ZFC* sample, obtained at $T = 15$ K by sweeping H_0 from 0 up to 10 kOe and back. By analyzing the experimental data at low H_0 values, one notes that R_s starts to vary at H_0 lower than the expected value of the first-penetration field of Abrikosov fluxons. This behavior is consistent with the results of the *AC* susceptibility and shows that also at *mw* frequency the presence of weak links significantly contributes to the energy losses. Moreover, one can see a clockwise magnetic hysteresis similarly to what observed in the $\chi'(H_0)$ curve.

Magnetic hysteresis in the *mw* surface resistance of type-II *SC* in the mixed state has been detected by different authors [11, 16, 17]; it has been ascribed to the different value of the magnetic induction inside the sample at increasing and decreasing *DC* fields, due to the critical state of the fluxon lattice. In Refs. [11, 13], we have investigated the effect of the critical state of the Abrikosov-fluxon lattice in $R_s(H_0)$ and have quantitatively described the hysteretic behavior of $R_s(H_0)$. Since the field-induced variations of R_s are related to the presence and motion of fluxons, it is expected that the shape of the hysteresis is strongly related to that of the magnetization curve, giving rise to a higher value of R_s in the decreasing-field branch with respect to that of the increasing-field branch. So, taking into account the effects of the critical state of the Abrikosov-fluxon lattice, a clockwise hysteresis cannot be explained. Another result of our investigation is that the amplitude of the hysteresis is proportional to the full penetration field, H^*: samples of small size and/or small J_c are expected to exhibit weak hysteretic behavior.

Clockwise hysteresis in $R_s(H_0)$ has been detected in granular cuprate *SC* and has been quantitatively discussed by Ji et *al.* [17] in terms of the so-called two-level critical-state model, which considers a large critical current density inside the grains, J_{cg}, and a much weaker J_{cm}. The authors calculate the intergrain fluxon density and show that the contribution to the *mw* losses of the intergrain fluxons accounts for the clockwise hysteresis of $R_s(H_0)$.

We would like to remark that the model elaborated by Ji et *al.* for explaining the feature of the $R_s(H_0)$ curve and those [4, 6] we have discussed in Sec. 3.1 to account for the hysteretic behavior of $\chi(H_0)$, assume that the main contribution responsible for the in-field *em* response comes from the intergrain fluxon dynamics. In principle, in order to calculate the field-induced variation of R_s one should take into account two contributions; one arising from critical state of intragrain fluxons, which should give rise to a counterclockwise hysteresis, and another arising from critical state of intergrain fluxons, which should give rise to a clockwise hysteresis. Our results suggest that for *DC* fields up to 10 kOe the intragrain contribution in this sample is negligible. Two different reasons may be responsible of the negligible intragrain contribution to the hysteresis of $R_s(H_0)$: i) the applied magnetic field is much lower than H_{c2} with consequent low fluxon density in the grains; ii) the grains have small H^* because of their small size with consequent small trapped flux.

4. Conclusion

We have investigated the low and high frequency response of a $LaO_{0.94}F_{0.06}FeAs$ sample exposed to DC magnetic fields up to 10 kOe, by measuring the AC susceptibility at 100 kHz and the microwave surface resistance at 9.6 GHz. The results have been qualitatively discussed in the framework of models reported in the literature. We have shown that the *em* response is strongly affected by the granular nature of the sample. Both at zero and at high DC magnetic fields, we have highlighted nonlinear effects strictly related to the critical state of fluxons in the intergrain region.

Measurements of the susceptibility at zero DC magnetic field as a function of the AC driving field have highlighted values of the intergrain full-penetration field smaller than 1 Oe, even at the lowest temperature investigated. This very small value of H_m^*, although further reduced by magnetic-relaxation during the AC cycle, indicates extremely low pinning potential. At DC magnetic fields higher than 100 Oe, both χ' and χ'' show a magnetic hysteresis, which can be justified considering the intragrain-field-penetration effects on the intergrain critical current density.

The field dependence of the microwave surface resistance exhibits a clockwise hysteresis, similar to that detected for χ', which cannot be justified in the framework of critical-state models of the Abrikosov-fluxon lattice. According to the models reported in the literature for granular superconductors, the clockwise magnetic hysteresis of R_s can be accounted for by supposing that the main contribution to the field-induced *mw* losses comes from the intergrain region.

Acknowledgments

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